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DIGITAL REFRACTOMETRY OF PIEZOELECTRIC CRYSTALLINE MEDIA

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The current report is an Interim Report for an SBIR Phase II award and commenced on August 1, 1987. The contractual objectives of this program are as follows:

1. Perform a theoretical analysis of the measurement of piezoelectric crystals using Dual-Beam Digital Refractometry,
2. Set up the laboratory configuration to determine the refractive indices of piezoelectric crystals using a HeNe Laser source,
3. Determine the refractive indices of various piezoelectric crystals,
4. Expand the measurements to the ultraviolet and infrared regions,
5. Analyze the requirements for applying Dual-Beam Digital Refractometry to the millimeter and the microwave regions.

Contract objective 1 has been completed and is now being written up as a publication. Contract objective 2 was completed by the end of August, 1988. Measurements are now (contd)

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19. ABSTRACT (contd)

being performed on various samples and the first results will be submitted by the end of February, 1989. Contract objectives 3, 4, and 5 will be completed at the end of the contract, February, 1990.

TABLE OF CONTENTS

1. Introduction.

The current report is an Interim Report for an SBIR Phase II award, "Digital Refractometry of Piezoelectric Crystalline Media", DAAL01-87-C-0727, and commenced on August 1, 1987.

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Contract objective 1. has been completed and is now being written up as a publication and will be submitted by the end of February, 1989. Contract objective 2. was completed by the end of August, 1988. Measurements are now being performed on various samples and the first results will be submitted by the end of February, 1989. Contract objectives 3., 4. and 5. will be completed at the end of the contract, February, 1990.

We now present a brief overview of Digital Refractometry along with the results to date.

The fundamental parameters of optical materials are the refractive index and the absorption coefficient. In this phase of the program we restricted ourselves to determining only the refractive index of optical materials using a new measurement method called Digital Refractometry. There are several classical methods for measuring the refractive index. The best known are 1)the minimum deviation prism method and 2)the Brewster angle null method. The former is a transmission method and the latter is a reflection method. The use of either method has advantages and disadvantages; these have been discussed in earlier reports and will not be repeated here. Needless to say, however, both methods require very expensive mechanical rotating mounts in order to collect the data. Furthermore, both methods are surprisingly difficult and expensive to automate. These methods can be characterized as being opto-mechanical. That is, optical components and mechanical components are necessary to measure the refractive index. The data is collected mechanically by moving the divided mechanical circle to the minimum deviation for the case of the minimum deviation prism method or to the null intensity position for the Brewster angle method. It is not difficult to understand that automating a mechanical circle to make arc second measurements is difficult and expensive.

In order to overcome these limitations Measurement Concepts, Inc. developed a new method for measuring the refractive index called Digital Refractometry. In contrast to the opto-mechanical methods Digital Refractometry is an opto-electronic method. That is, the data are collected optically and electronically. Furthermore, unlike the opto-mechanical methods, Digital Refractometry can be made to operate with no moving mechanical parts. Digital Refractometry rests on a little-known simplification in Fresnel's reflection equation at an incident angle of 45° and the existence of high resolution digital voltmeters. While Fresnel's equations have, of course, been known for a long time there was very little motivation for investigating their form at an incident angle of 45^{o} until recently. In fact, it appears that until 1960 Fresnel's equations and the corresponding Mueller matrices were known to reduce to simple forms only for normal incidence and the Brewster angle. A study of the literature has revealed that the simplification in Fresnel's equations at an incidence angle of 45^{o} was only realized in 1960 by the English physicist, Sir Humpreys-Owen. However, this simplification would not by itself be very useful. In order to use this simplification, practically, it is necessary to measure the reflected intensities digitally, that is, with digital voltmeters. Digital voltmeters only became available during the 1970s so this type of measurement could not be carried out earlier.

In Phase I of this SBIR program we showed that it was possible to determine the refractive index to three decimal places with a 4&1/2 digital voltmeter. Analyses and experiments revealed that, in general, it is possible to measure the refractive index to m places with a (m+1)&1/2 digital voltmeter. For example, the refractive index can be read to 5 decimal places with a 6&1/2 digital voltmeter. Another important result of the analysis is that only two orthogonal intensity measurements, I_s and I_p, need be made to determine the refractive index. These are the perpendicular (s) polarization intensity, I_s, and the parallel (p) polarization intensity, I_p, respectively. Furthermore, the ratio of I_p/I_s is of the order of 10:1 so the dynamic range of a system need only be 10dB.

In this first half of the Phase II SBIR program our major objective was to design and implement a laboratory optical system for Digital

Refractometry capable of measuring the refractive index to at least five decimal places. This required that we purchase an amplitude stable HeNe laser, two lock-in amplifiers, two 6&1/2 digital voltmeters and automated rotational mounts capable of moving to within an arc second under an electronic controller; we also required the highest quality Glan-Thompson calcite polarizers. In principle, it is not necessary to use rotational mounts to implement Digital Refractometry. However, for initial alignment and data collection using Fourier methods it is very desirable to have them.

By the end of August, 1988 the major pieces of equipment purchased were received. The laboratory has now been completely assembled. Since September work has proceeded on measuring the refractive index of selected samples to five decimal places. Surprisingly, we have found that we have very "slow", ac-like, fluctuations throughout our system. This fluctuation has prevented us from making fifth-place measurements on a consistent basis. When the system was "turned on" in late August it was thought, initially, that this was due only to laser fluctuations and would be eliminated by using a ratiometer. Hence, a ratiometer was purchased. Again, to our surprise, the fluctuations did not disappear even with the use of the ratiometer. We are fairly certain that we have an "incorrect" ground in our system. Furthermore, we have discovered that we obtain very stable, fifth place, readings late in the evening when the surroundings have "quieted down". We have ordered a number of pieces of test equipment in order to determine the cause of these fluctuations. We expect that this problem will be solved by the end of December, 1988, whereupon the measurement program can proceed.

2. Technical Discussion.

The fundamental concept of Digital Refractometry was presented in our Phase I SBIR Final Report as well as our Phase II SBIR Proposal. We briefly review the basic idea behind Digital Refractomety.

Digital Refractometry was developed to determine the refractive index of optical materials to five and six decimal places. It rests on a little known simplification in Fresnel's equations at an incidence angle of 45^{o} and the existence of high resolution digital voltmeters. In principle, Digital Refractometry can be made to work without any moving mechanical parts; the method is then spoken as being opto-electronic. That is, using only optics and electronics the refractive index can be measured. This is a significant shift from the current methods for measuring the refractive index which are the minimum deviation prism method and the Brewster angle null method; these methods are opto-mechanical. That is, mechanical movement with a divided circle is required to make the measurement.

The simplest configuration for implementing Digital Refractometry is shown in Fig.1. In our experiment a Spectra-Physics 117 Amplitude Stable HeNe Laser is used. Measurements made by us have shown that this laser is stable to nearly 1 part in 10,000, which is excellent. The laser beam then propagates through a calcite Glan-Thompson linear polarizer with its transmission axis set at 45^{o}. The polarizer is mounted in a Newport rotational mount 495A and is capable of being rotated to within 0.001^{o} (3.6 arc secs). Three of these mounts are used in the current experimental configuration. The first is used to generate the 45^{o} linearly polarized light, the second is used to rotate the sample to either 45^{o} in the plane of optical table or to the

Brewster angle and the third rotational mount contains another linear calcite polarizer for analyzing the reflected optical beam. All three mounts are motorized and are under the control of a Newport Controller 850; it has performed extremely well and operates within the specifications.

In the measurements an optically isotropic standard BK7 2" glass disk is mounted in a Newport 610 ultraresolution mount; it is capable of being aligned to within 1 arc sec. The reflected beam is then analyzed by a linear calcite polarizer. Both calcite polarizers were made under special order by Continental Optics, Farmingdale, Long Island, from the same piece of calcite. Measurements have shown that when the polarizers are crossed the intensity read on the 6&1/2 Keithley 196 digital voltmeter is 0.000001 which is practically perfect. Analysis shows that the refractive index can be determined by simply measuring the "s" polarized intensity, I_s, and the "p" polarized intensity, I_p, respectively. The refractive index can then be shown to be given by

$$n = \frac{\sqrt{(I_s + I_p)}}{\sqrt{I_s} - \sqrt{I_p}} \quad . \tag{1}$$

A rigorous derivation of this formula has been given by A. Ballato. An alternative derivation, albeit less rigorous but simpler, and based on symmetry arguments can also been made and is presented in Appendix I.

It is of interest to determine the resolution requirement of a digital voltmeter to determine the refractive index, say, to five decimal places. Ballato's formula can be solved for R and is found to be

$$R = [n^2 + \surd(2n^2 - 1)]^2/(n^2 - 1)^2 \ . \quad (2)$$

where $R = I_s/I_p$. At 6328Å the refractive index of BK7 is n=1.51509 according to the Schott optics catalog. The maximum resolution of a 6&1/2 digital voltmeter is 1.999999 volts; it is sufficient to write this, for numerical convenience, as 2.000000 volts or simply 2. If I_s is set to 2.000000 volts then $I_p = I_s/R = 2/R$. The following table has been constructed for BK7 and shows the resolution requirement on I_p in order to measure the refractive index to five and six decimal places:

n	R	I_p(5 places)	I_p(6 places)
1.51508	10.46326	0.19115	0.191145
1.51509	10.46300	0.19115	0.191150
1.51510	10.46274	0.19115	0.191154

Thus, in order to measure a difference in n in the fifth decimal place it is necessary to measure I_p to the sixth decimal place. That is, a 6&1/2 digital voltmeter is required.

It is also necessary to know the resolution requirements of the analyzing polarizer and its effect on the required resolution of the digital voltmeter. A straightforward calculation shows that if the angle of refraction is r and the transmission axis of the polarizer is set at θ then the intensity on the detector will be

$$I(\theta) = [(1-\sin 2r)/(1+\sin 2r)^2][1+\sin(2r-2\theta)]. \qquad (3)$$

Eq.(3) can be written simply as

$$I(\theta) = I_0[1+\sin(2r-2\theta)] \quad . \tag{4}$$

Then

$$\Delta I/I = [-2\cos(2r-2\theta)/(1+\sin(2r-2\theta)]\Delta\theta \quad . \tag{5}$$

We are capable of measuring $\Delta I/I = 0.000001/2$ with a 6&1/2 DVM. Assume that $2r-2\theta = 135^{o}$. Then,

$$\Delta\theta = 0.6X10^{-6}\text{rads} = 0.1 \text{ arc secs} \tag{6}$$

This is far below the value of 3.6 arc secs (0.001^{o}) of the Newport mounts. In fact, a quick calculation using $\Delta\theta = 0.001^{o}$ shows that the corresponding value of $\Delta I/I$ is 0.000014. Thus, at this time we cannot do better than 5 decimal places on the DVMs using rotational mounts. However, in a final configuration where we do not use rotational mounts and the polarizers are fixed it should be possible to attain the value predicted by Eq.(6) using a new method we have developed. We now discuss this method.

The alignment of the generating and analyzer polarizers must be parallel to the optical table. That is, the transmission axes of the polarizers must be 0^{o} in the parallel postion. Traditionally, this is done by using a Brewster angle method. However, this has the disadvantage of requiring that we move to the sample to the Brewster angle; this can only be done to 0.001^{o} in the horizontal plane and, similarly, in the vertical plane. Consequently, we have derived an alternative method to determining the horizontal axis. As is well known when linear polarizers are crossed there will be a null. However, this is a relative measurement because the transmission axis of one polarizer can be at θ. It is only necessary to rotate the analyzer to $\theta+90^{o}$ to obtain the null condition. Our problem is to be certain that θ is actually zero. This can be done by aligning the sample (which is set at 45^{o}) with the autocollimator; using the autocollimator assures us that the sample is within 1 arc second and the normal of the sample is parallel to the plane of the optical table. Under this condition the Mueller matrix for the generator, polarizing sample and analyzer will be

$$M = \begin{pmatrix} 1 & -1 & 0 & 0 \\ -1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} A & B & 0 & 0 \\ B & A & 0 & 0 \\ 0 & 0 & C & 0 \\ 0 & 0 & 0 & C \end{pmatrix} \begin{pmatrix} 1 & 1 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \tag{7}$$

where we have dropped the factor of 1/4 in front of the matrices. Carrying out the matrix multiplication we obtain $M = 0$. Thus, by using the sample we can align the axes of the polarizers parallel and perpendicular to the optical table. We have used this procedure with great success. This procedure will be described completely in the final report.

Two other methods have also been used by us to determine the refractive index, n. Inspection of Eq.(4), above, shows that will obtain a null intensity at $2r-2\theta=270^{o}$ or $r-\theta=135^{o}$ and $r=135^{o}-\theta$. This method has also been used and leads to values in very good agreement with the orthogonal intensity method we have been using to determine the refractive index. It too will be described in detail in the final report.

Finally, it is possible to do a least-means-squares data fit by doing a Fourier analysis on Eq.(4). To show this we write Eq.(4) as

$$I(\theta)=I_0[1 + \sin 2r\cos\theta - \cos 2r\sin\theta] . \qquad (8)$$

A straightforward Fourier analysis of Eq.(8) then yields

$$I_0 = (1/N)\sum I_n , \qquad (9a)$$

$$\sin 2r = (2/N)\sum I_n\cos 2\theta_n , \qquad (9b)$$

$$\cos 2r = -(2/N)\sum I_n\sin 2\theta_n . \qquad (9c)$$

A computer program has been written to read every 0.1 degree over 360° (3,600 data points). However, because of the difficulties we have experienced with the intensity fluctuations we have not carried out this method. We expect that with the elimination of the fluctuations we shall be able to this.

In summary, the laboratory is now complete and we expect to take fifth and sixth place data by January, 1989. At this writing, which is mid-December we have already reduced the amount of fluctuations by replacing all the cables, checking the ground connections and tightening all the screws and cables, grounding the optical table; we have also ordered battery operated transimpedance amplifiers, a Tektronix 2236 oscilloscope and a battery operated Keithley 175 DVM.

3. Conclusions.

At the beginning of September, 1988 the laboratory configuration for measuring the refractive indices of piezoelectric crystals was completed. All of the individual pieces of equipment ordered by us were tested and performed correctly. However, when the pieces were integrated as a system a persistent small ac fluctuation appeared in the fourth and fifth decimal place on the digital voltmeters. We have continued to investigate the cause of these fluctuations and to try to correct them. We believe that with the new monitoring equipment such as the purchase of a Tektronix 2236 100MHz oscilloscope we shall be able to determine the nature of these fluctuations. We have purchased two UDT 101 battery operated transimpedance amplifiers in order to isolate these units from the observed fluctuations. In addition, we have purchased a battery operated Keithley 175 4&1/2 digital voltmeter for making initial measurements and to determine if the ac fluctuations can be eliminated. These items have been ordered and will arrive in December.

The software for measuring the refractive index automatically has been completed and checked. However, because we have not succeeded at this date in overcoming the fluctuation problem we have not run it totally on the system. To date it has been run as subprograms, that is, automated rotation of the mounts, Brewster angle measurements, data collection and data reduction; in the beginning of next year the program for doing a Fourier analysis shall be written; the control and data collection are done under a GPIB interface.

We are also building a temperature control for the piezoelectric crystal mount. A first version was completed in October and worked reasonably well. However, several mechanical improvements were needed and these are now being made. We expect to have these improvements completed by February, 1989.

In summary, all the apparatus for measuring the refractive index to at least five decimal places is now in place. During the month of December we shall track down the source of the voltage fluctuations and correct them. This activity will be completely described in the monthly report for December. It is expected that by the end of December the fluctuation problems will be solved and corrected and the measurement program can then proceed.

APPENDIX I. RELATION BETWEEN THE REFRACTIVE INDEX AND THE REFLECTION RATIO.

A very useful relation has been found by A. Ballato between the refractive index, n, and the reflection ratio, $R=I_s/I_p$. Namely,

$$n = \frac{\sqrt{(I_s + I_p)}}{\sqrt{I_s} - \sqrt{I_p}} \quad . \qquad \text{(I-1)}$$

An alternate derivation of this formula is now presented. The following relation was derived earlier relating the angle of refraction, r, to the measured intensities I_s and I_p,

$$\sin 2r = (I_s - I_p)/(I_s + I_p) \quad . \qquad \text{(I-2)}$$

Eq.(I-2) can be written as

$$2\sin r\cos r = (R - 1)/(R + 1) \qquad \text{(I-3)}$$

where $R=I_s/I_p$ is called the reflection ratio. Eq.(I-3) can now be expressed as

$$2\sin r\cos r = [(\sqrt{R} - 1)(\sqrt{R} + 1)]/[\sqrt{(R + 1)}\sqrt{(R + 1)}] \quad . \qquad \text{(I-4)}$$

The form of Eq.(I-4) suggests that it be factored in the following way,

$$\sqrt{2}\sin r = (\sqrt{R} - 1)/\sqrt{(R + 1)} \ , \ \sqrt{2}\cos r = (\sqrt{R} + 1)/\sqrt{(R + 1)} . \qquad \text{(I-5)}$$

In Digital Refractometry the incidence angle is 45°. For this condition Snell's law becomes

$$1/\sqrt{2} = n\sin r \tag{I-6}$$

so

$$n = 1/(\sqrt{2}\sin r). \tag{I-7}$$

Using Eq.(I-7) with Eq.(I-5) we find that

$$n = [\sqrt{(R + 1)}]/[\sqrt{R} - 1)] \tag{I-8}$$

which is Ballato's formula.

Eq.(I-5) satisfies the relation

$$\cos^2 r + \sin^2 r = 1 \tag{I-9}$$

and so is a satisfactory decomposition, although, not necessarily unique. A rigorous solution of Eq.(II-4) can be found by writing

$$2\sin r\cos r = 2\sin r\sqrt{(1 - \sin^2 r)} = f \tag{I-10}$$

where $f = (R-1)/(R+1)$ and then solving for $\sin r$. However, some additional algebraic factoring is still needed to bring the result into Ballato's formula.

Appendix II. Digital Refractometry for Uniaxial Crystals.

In order to apply Digital Refractometry to uniaxial as well as piezoelectric crystals it is necessary to know the appropriate forms of Fresnel's equations. While Fresnel's equations for reflection are well known for isotropic media the same cannot be same for Fresnel's equations for reflection from uniaxial (and uniaxial absorbing) crystals. In fact, only in 1984 were the equations published for uniaxial crystals suitable for experimental verification.

There is one experimental configuration which is of direct interest to Digital Refractometry; this is shown in Fig. 2. For this case the incident and reflected waves can be decomposed into two linearly polarized waves such that the following equations are valid:

$$E_z^r/E_z^i = [\cos\theta-(n_p^2-\sin^2\theta)^{1/2}]/[\cos\theta+(n_p^2-\sin^2\theta)^{1/2}] , \qquad \text{(II-1)}$$

$$E_{xy}^r/E_{xy}^i = [n_s^2\cos\theta-(n_s^2-\sin^2\theta)^{1/2}]/[n_s^2\cos\theta+(n_s^2-\sin^2\theta)^{1/2}] , \qquad \text{(II-2)}$$

where n_p and n_s are the refractive indices for the extraordinary and ordinary refractive indices, respectively. We see that these equations are not coupled so they provide unique solutions. For Digital Refractometry the incident angle is 45°. We then find from the above equations

$$E_z^r/E_z^i = -[(2n_p^2-1)^{1/2}-1]/[(2n_p^2-1)^{1/2}+1] \qquad \text{(II-3)}$$

and

$$E^r_{xy}/E^r_{xy} = -[(2n_s^2-1)^{1/2}-n_s^2]/[(2n_s^2-1)^{1/2}+n_s^2] \qquad \text{(II-4)}$$

Eqs.(II-3) and (II-4) are the fundamental pair of equations which are used in the application of Digital Refractometry to uniaxial (piezoelectric) crystals.

Eqs.(II-3) and (II-4) can be transformed to the Mueller matrix form, thereby, characterizing the piezoelectric crystal in the following way. We can write these equations, respectively as

$$E^r_z = aE^i_z \quad , \quad E^r_{xy} = bE^i_{xy} \qquad \text{(II-5)}$$

where a and b are the expressions involving the refractive indices, Eqs.(II-3) and (II-4). From the usual definition of the Stokes parameters for the incident and the reflected beam we then find that the Mueller matrix has the form

$$M = \begin{pmatrix} a^2 + b^2 & a^2 - b^2 & 0 & 0 \\ a^2 - b^2 & a^2 + b^2 & 0 & 0 \\ 0 & 0 & 2ab & 0 \\ 0 & 0 & 0 & 2ab \end{pmatrix} . \qquad \text{(II-6)}$$

Eq.(II-6) is a matrix of a polarizer. Its complete development will be presented in the Final Report.

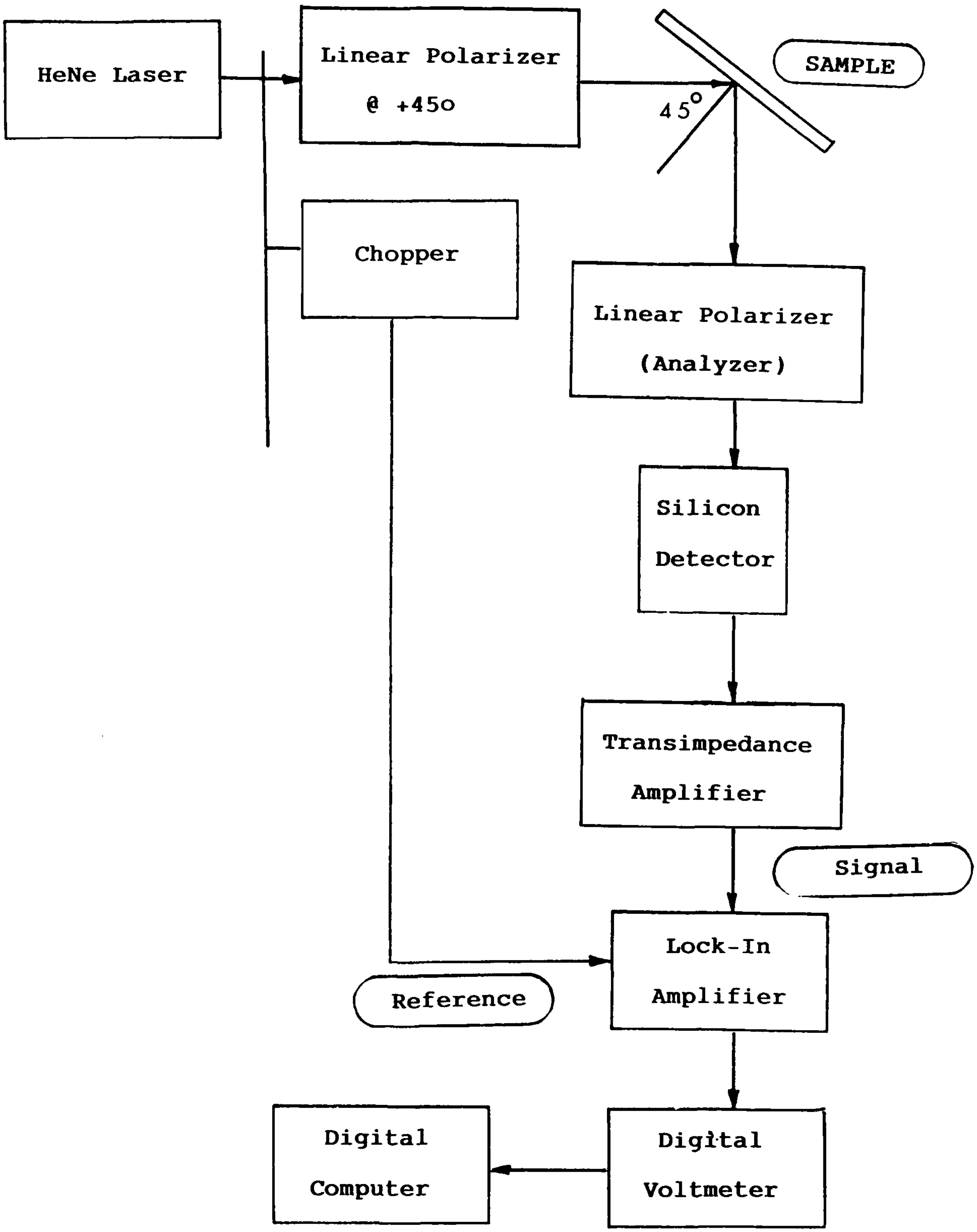

Fig. 1. Measurement configuration for Digital Refractometry.

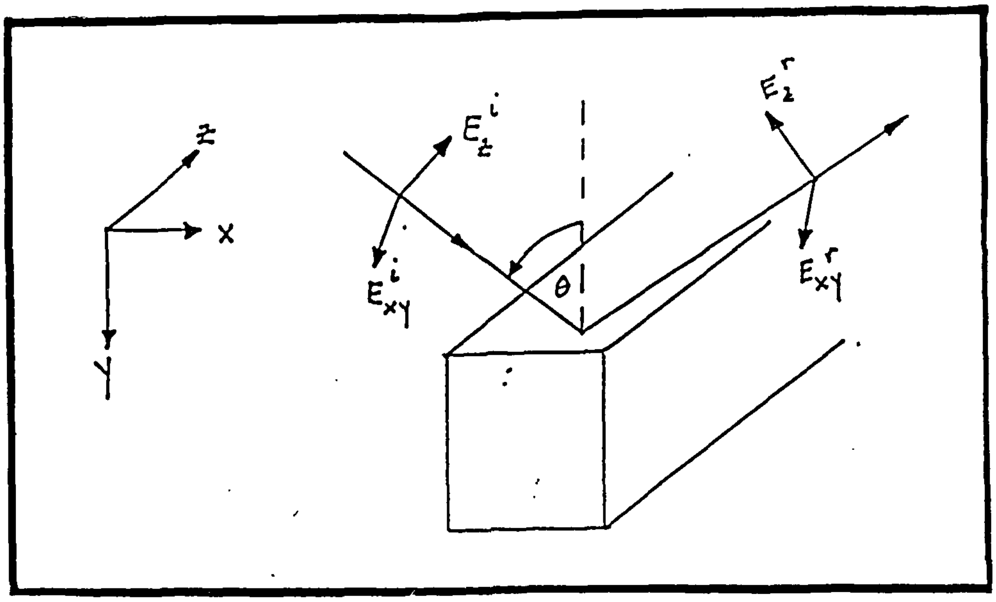

Fig. 2. Reflectance from a plane parallel to the optics axis with the plane of incidence parallel to the basal (the x-y plane). The optic axis is along the z axis.

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